



Spectroscopy for development of plasma sources for water window imaging

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ISCA Japan
International
Strategic
Cooperation
Award



Outline

Background:

Basics of laser produced plasmas, unresolved transition arrays.

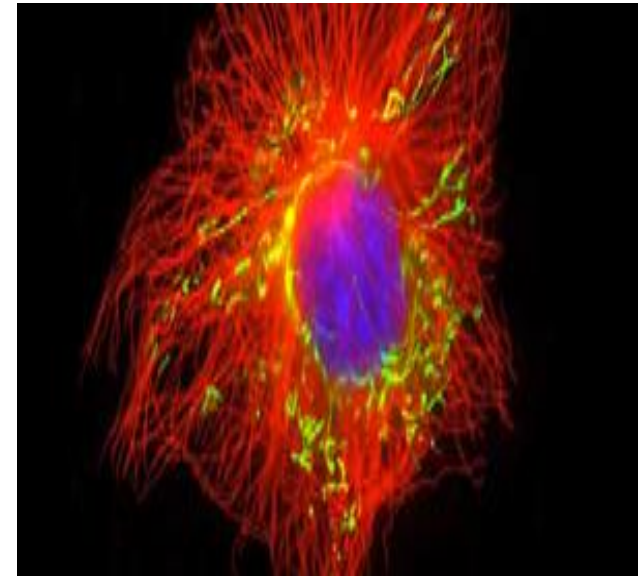
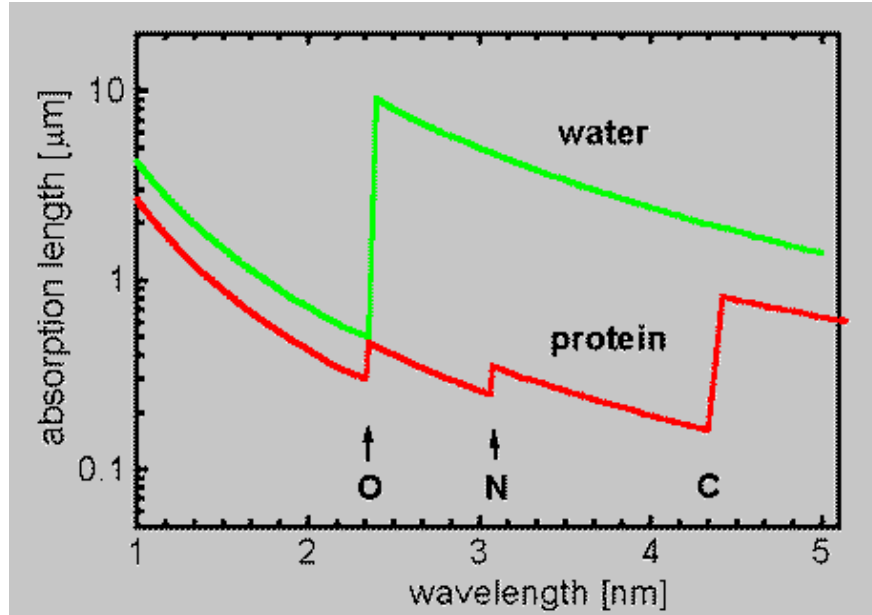
Spectroscopy:

$\Delta n=0$ Unresolved Transition Arrays

$\Delta n=1$ Unresolved Transition Arrays

Future Plans and Conclusions

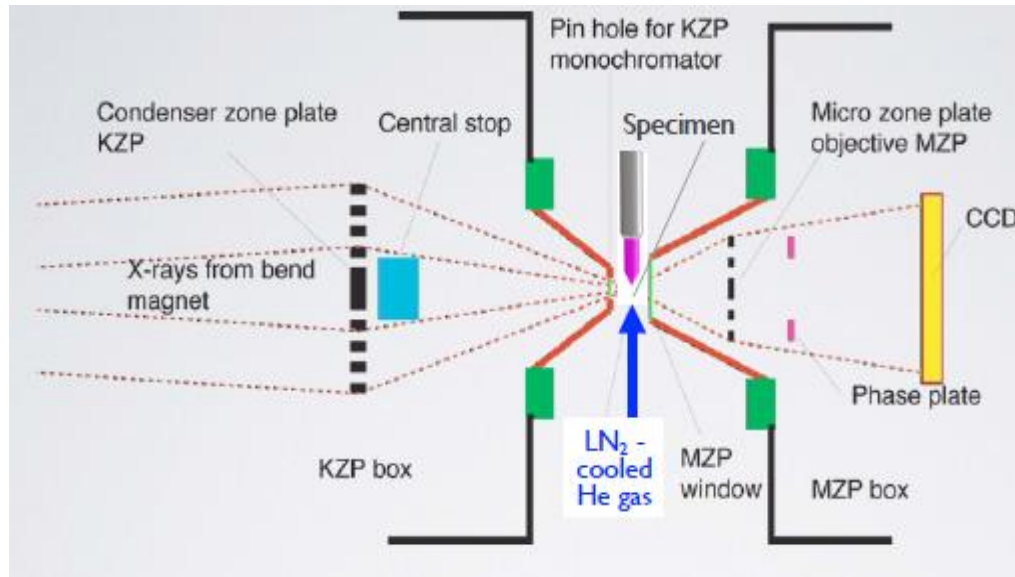
Water window



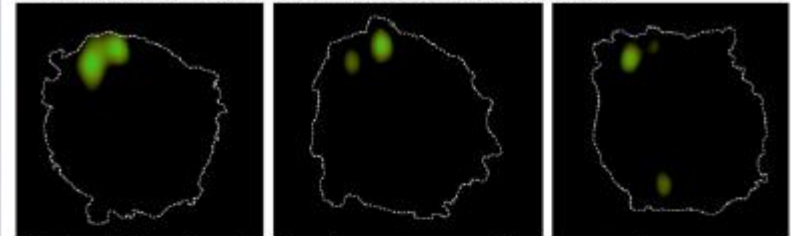
Water window : 2.34-4.38 nm.

Relative transparency of water allows investigation of biomolecules, cells and proteins in their natural aqueous environment by x-ray transmission microscopy or tomography

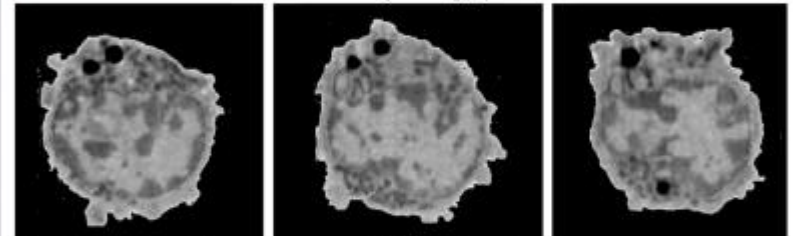
SXR Tomography: Zone Plate Optics



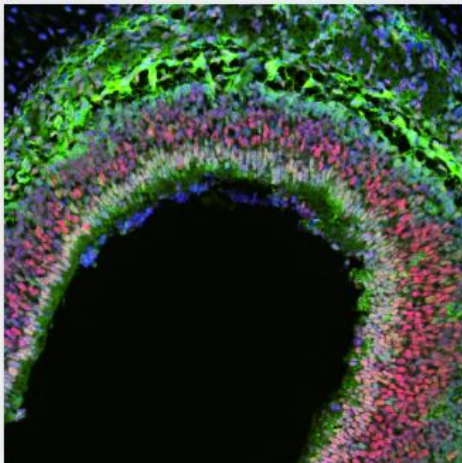
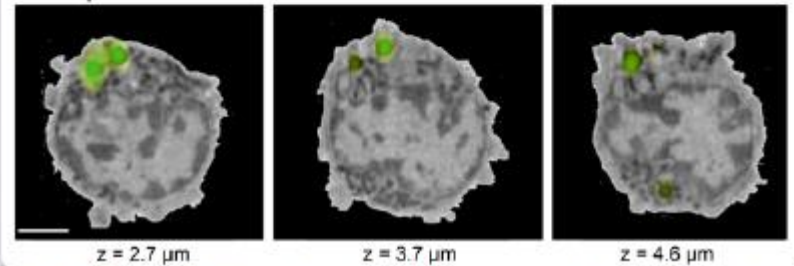
2D virtual slices from 3d cryogenic fluorescence tomography



2D virtual slices from the 3D soft x-ray tomographic reconstruction



Overlay of above fluorescence on SXT virtual sections



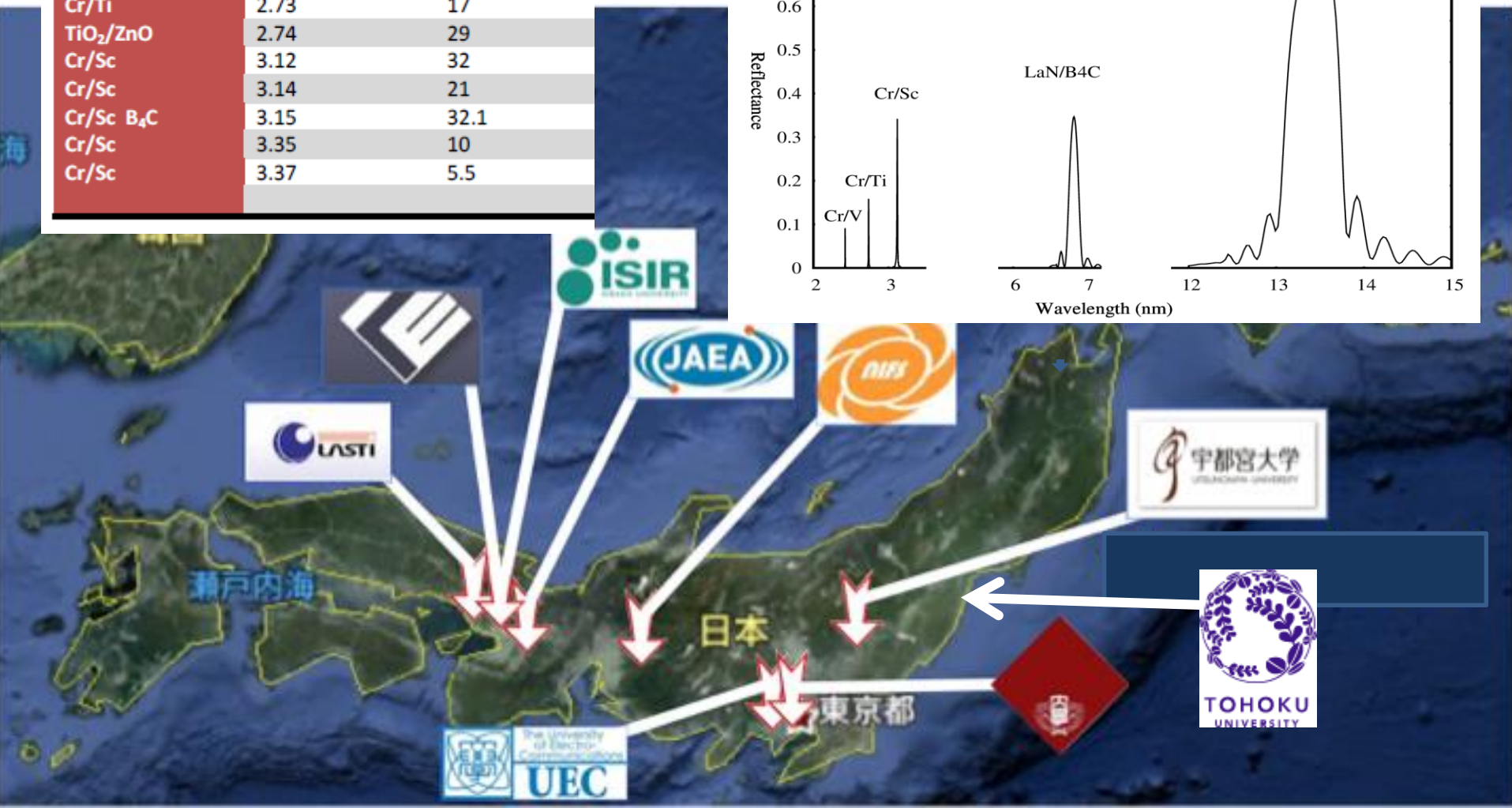
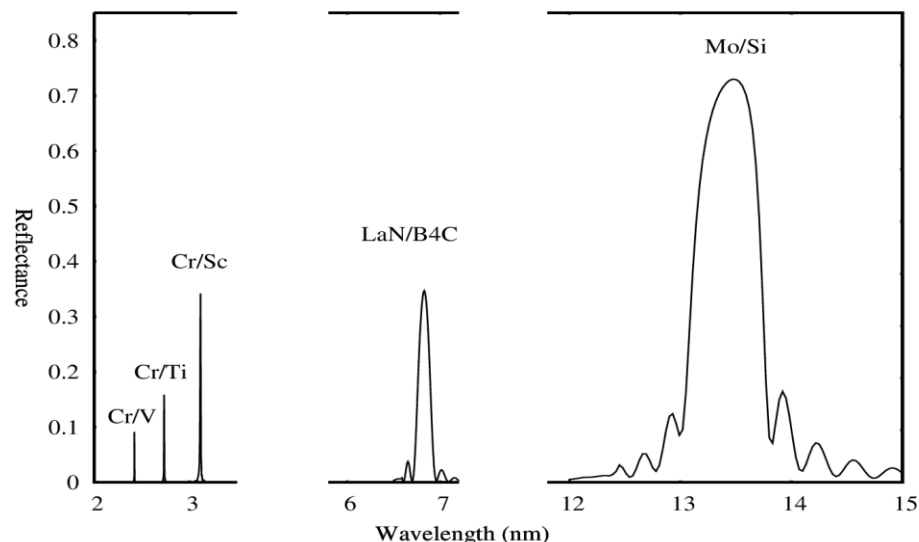
Stavros Lomvardas, UCSF

Smith EA, McDermott G, Do M, Leung K, Panning B, Le Gros MA and Larabell CA (2014). Quantitatively imaging chromosomes using correlated cryo-fluorescence and soft x-ray tomographies. *Biophysical Journal*. 107(8) 1988-96.

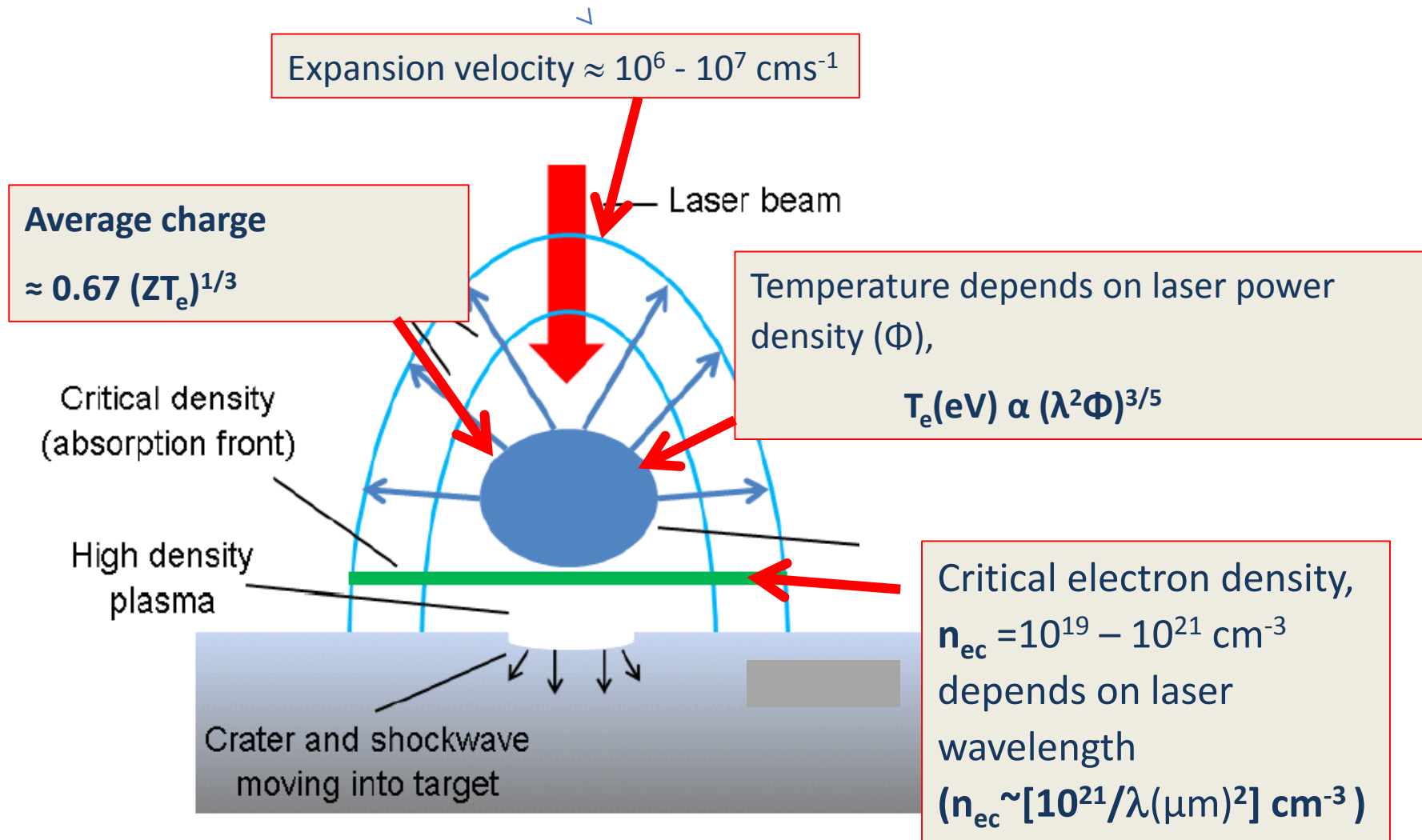
Carolyn Larabell, EUV Source Workshop, UCD S-3 2014

Network for single shot water window imaging source development with reflective optics

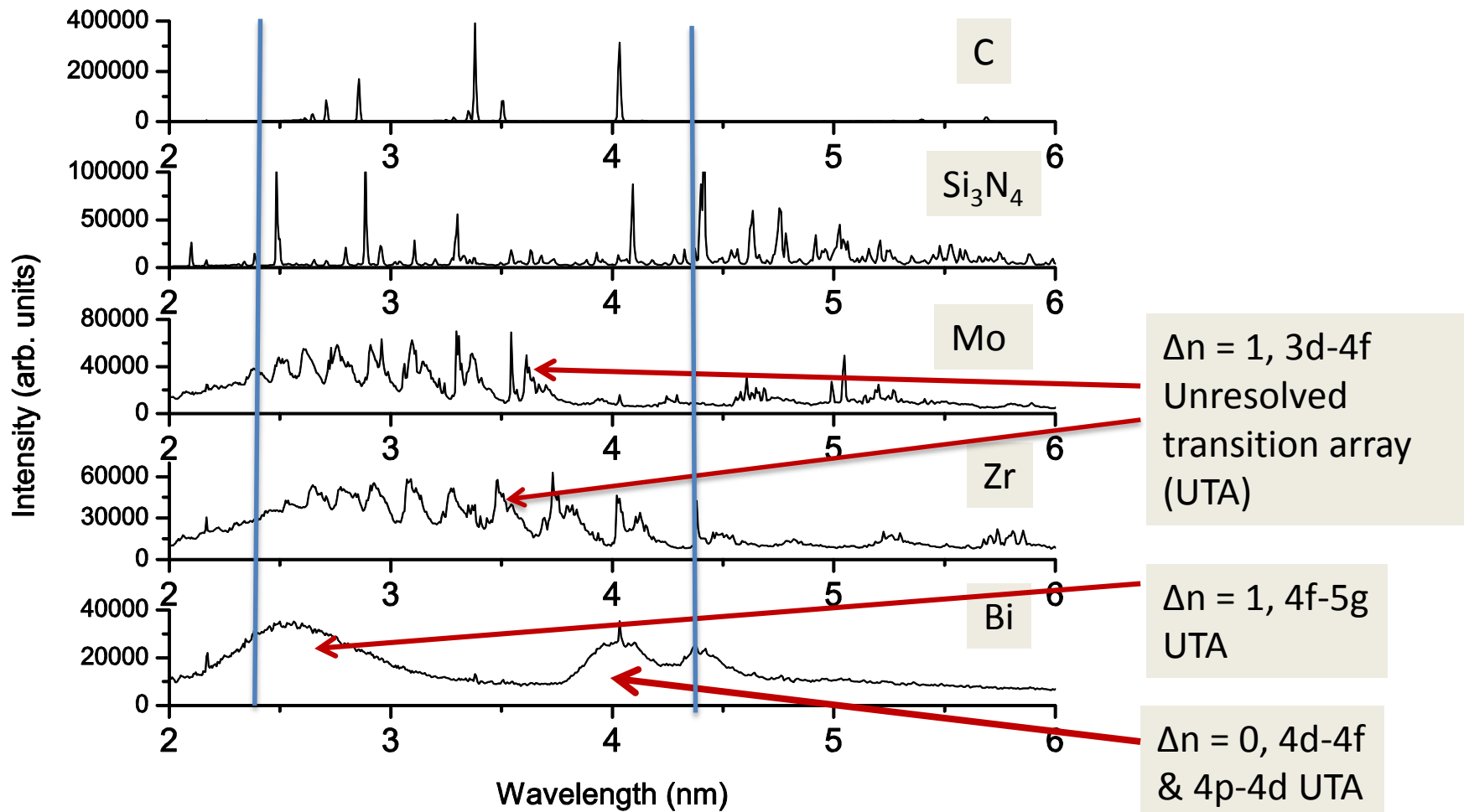
Material	Wavelength (nm)	Reflectivity (%)
Cr/V	2.42	9
Cr/Ti	2.73	17
TiO ₂ /ZnO	2.74	29
Cr/Sc	3.12	32
Cr/Sc	3.14	21
Cr/Sc B ₄ C	3.15	32.1
Cr/Sc	3.35	10
Cr/Sc	3.37	5.5



Laser produced plasmas (LPPs)



Comparison of water window emission spectra



Time-integrated spectra from picosecond-laser-produced plasmas

SXR spectra studied

1
1A
1A

2
2A
2A

3
3A
3A

4
4A
4A

5
5A
5A

6
6A
6A

7
7A
7A

8
8A
8A

9
9A
9A

10
10A
10A

11
11A
11A

12
12A
12A

13
13A
13A

14
14A
14A

15
15A
15A

16
16A
16A

17
17A
17A

18
18A
18A

1
H
Hydrogen
1.008

2
He
Helium
4.003

3
Li
Lithium
6.941

4
Be
Beryllium
9.012

5
B
Boron
10.811

6
C
Carbon
12.011

7
N
Nitrogen
14.007

8
O
Oxygen
15.999

9
F
Fluorine
18.998

10
Ne
Neon
20.180

11
Na
Sodium
22.990

12
Mg
Magnesium
24.305

13
Al
Aluminum
26.982

14
Si
Silicon
28.086

15
P
Phosphorus
30.974

16
S
Sulfur
32.066

17
Cl
Chlorine
35.453

18
Ar
Argon
39.948

19
K
Potassium
39.098

20
Ca
Calcium
40.078

21
Sc
Scandium
44.956

22
Ti
Titanium
47.88

23
V
Vanadium
50.942

24
Cr
Chromium
51.996

25
Mn
Manganese
54.938

26
Fe
Iron
55.933

27
Co
Cobalt
58.933

28
Ni
Nickel
58.693

29
Cu
Copper
63.546

30
Zn
Zinc
65.39

31
Ga
Gallium
69.732

32
Ge
Germanium
72.61

33
As
Arsenic
74.922

34
Se
Selenium
78.09

35
Br
Bromine
79.904

36
Kr
Krypton
84.80

37
Rb
Rubidium
84.468

38
Sr
Strontium
87.62

39
Y
Yttrium
88.906

40
Zr
Zirconium
91.224

41
Nb
Niobium
92.906

42
Mo
Molybdenum
95.94

43
Tc
Technetium
98.907

44
Ru
Ruthenium
101.07

45
Rh
Rhodium
102.906

46
Pd
Palladium
106.42

47
Ag
Silver
107.868

48
Cd
Cadmium
112.411

49
In
Indium
114.818

50
Sn
Tin
118.71

51
Sb
Antimony
121.760

52
Te
Tellurium
127.6

53
I
Iodine
126.904

54
Xe
Xenon
131.29

55
Cs
Cesium
132.905

56
Ba
Barium
137.327

57
La
Lanthanum
138.906

58
Ce
Cerium
140.115

59
Pr
Praseodymium
140.908

60
Nd
Neodymium
144.24

61
Pm
Promethium
144.913

62
Sm
Samarium
150.36

63
Eu
Europium
151.966

64
Gd
Gadolinium
157.25

65
Tb
Terbium
158.925

66
Dy
Dysprosium
162.50

67
Ho
Holmium
164.930

68
Er
Erbium
167.26

69
Tm
Thulium
168.934

70
Yb
Ytterbium
173.04

71
Lu
Lutetium
174.967

72
Hf
Hafnium
178.49

73
Ta
Tantalum
180.948

74
W
Tungsten
183.85

75
Re
Rhenium
186.207

76
Os
Osmium
190.23

77
Ir
Iridium
192.22

78
Pt
Platinum
195.08

79
Au
Gold
196.967

80
Hg
Mercury
200.59

81
Tl
Thallium
204.383

82
Pb
Lead
207.2

83
Bi
Bismuth
208.980

84
Po
Polonium
[209]

85
At
Astatine
209.987

86
Rn
Radon
222.018

87
Fr
Francium
223.020

88
Ra
Radium
226.025

89-103
Actinide Series

104
Rf
Rutherfordium
[261]

105
Db
Dubnium
[262]

106
Sg
Seaborgium
[266]

107
Bh
Bohrium
[264]

108
Hs
Hassium
[269]

109
Mt
Meitnerium
[268]

110
Ds
Darmstadtium
[269]

111
Rg
Roentgenium
[272]

112
Cn
Copernicium
[277]

113
Uut
Ununtrium
unknown

114
Fl
Flerovium
[289]

115
Uup
Ununpentium
unknown

116
Lv
Livermorium
[298]

117
Uus
Ununseptium
unknown

118
Uuo
Ununoctium
unknown

Atomic Number

Symbol

Name

Atomic Mass

Alkali Metal

Alkaline Earth

Transition Metal

Basic Metal

Semimetal

Nonmetal

Halogen

Noble Gas

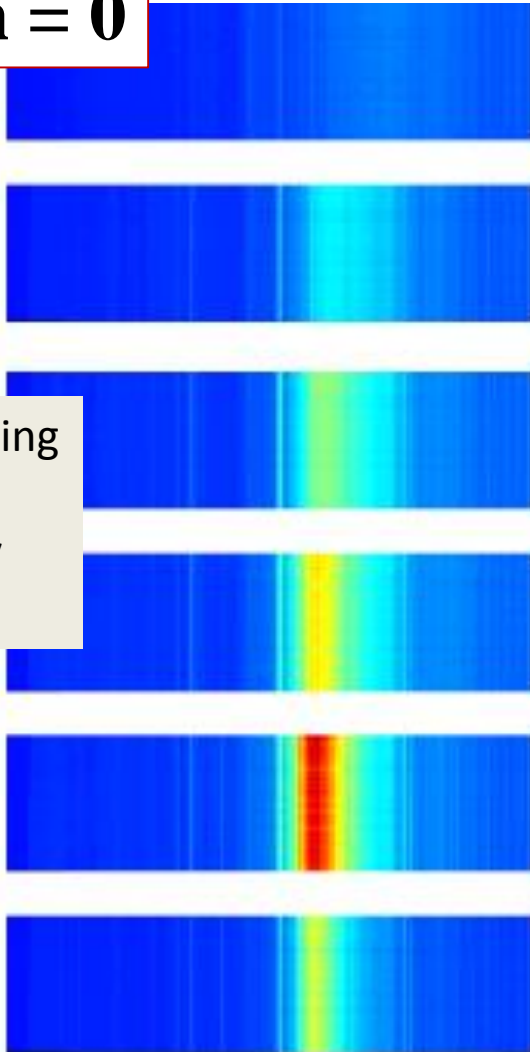
Lanthanide

Actinide

Two types of UTA in XUV spectra

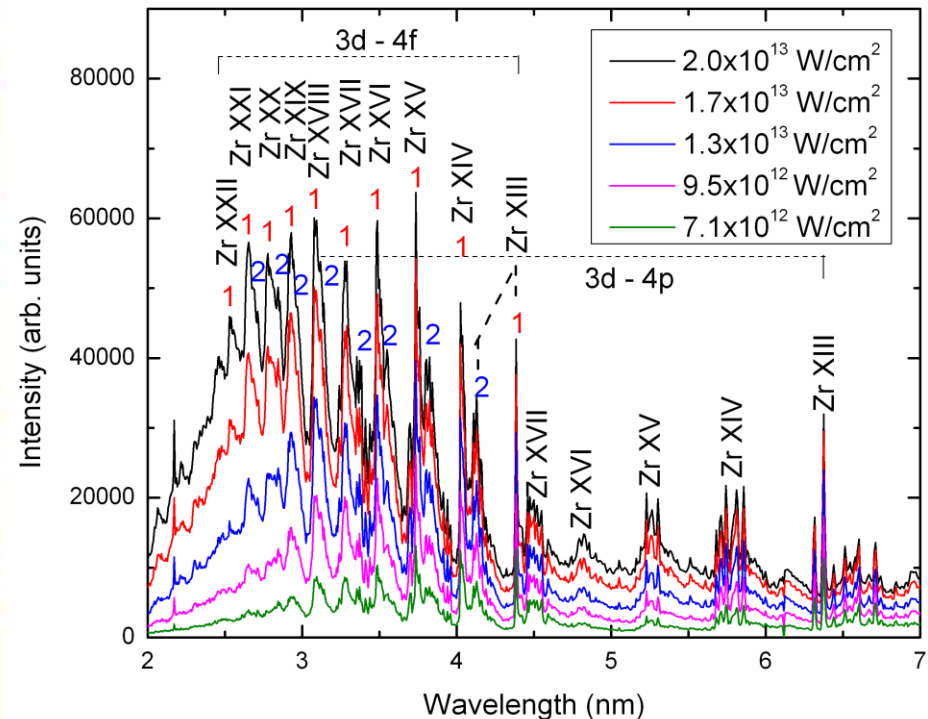
$$\Delta n = 0$$

Increasing
power
density
↓



$4p^6 4d^{N+1} - 4p^6 4d^N 4f + 4p^5 4d^{N+2}$
in Sn

$$\Delta n > 0$$



$\Delta n = 0$ transitions overlap in adjacent ion stages.

$\Delta n = 0$ transitions do not overlap in adjacent ion stages and move to shorter wavelengths with increasing ionization.

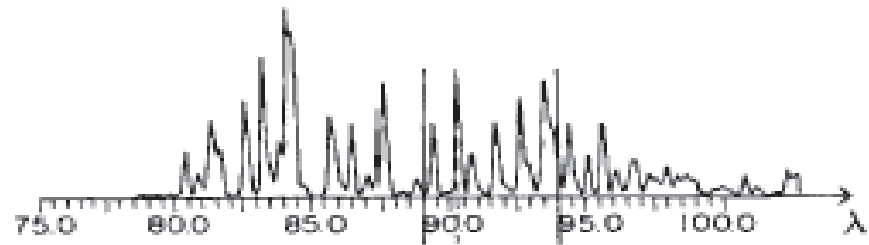
Spectral narrowing in mixed $\Delta n=0$ arrays

In spectra due to $4p^6 4d^{N+1} - 4p^6 4d^N 4f + 4p^5 4d^{N+2}$ transitions

Configuration interaction causes

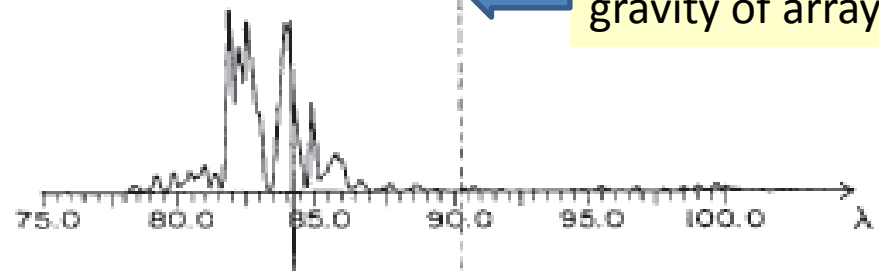
No CI

- Spectral narrowing.
- Strong peaking of oscillator strength.



Pr XXII: $4p^6 4d^2 - 4p^6 4d 4f + 4p^5 4d^3$

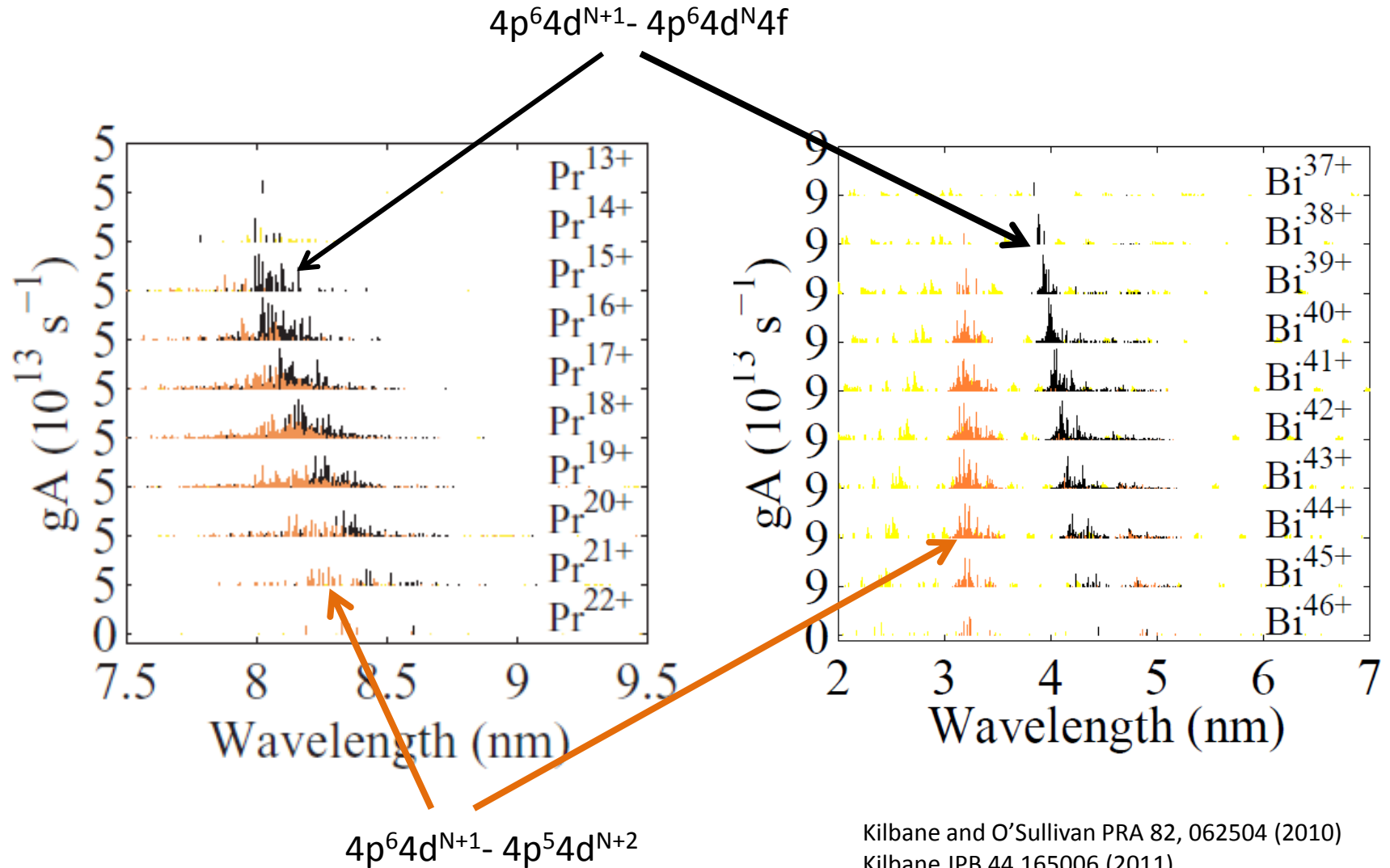
CI



Localisation of transitions at ~
the same position in successive
ion stages

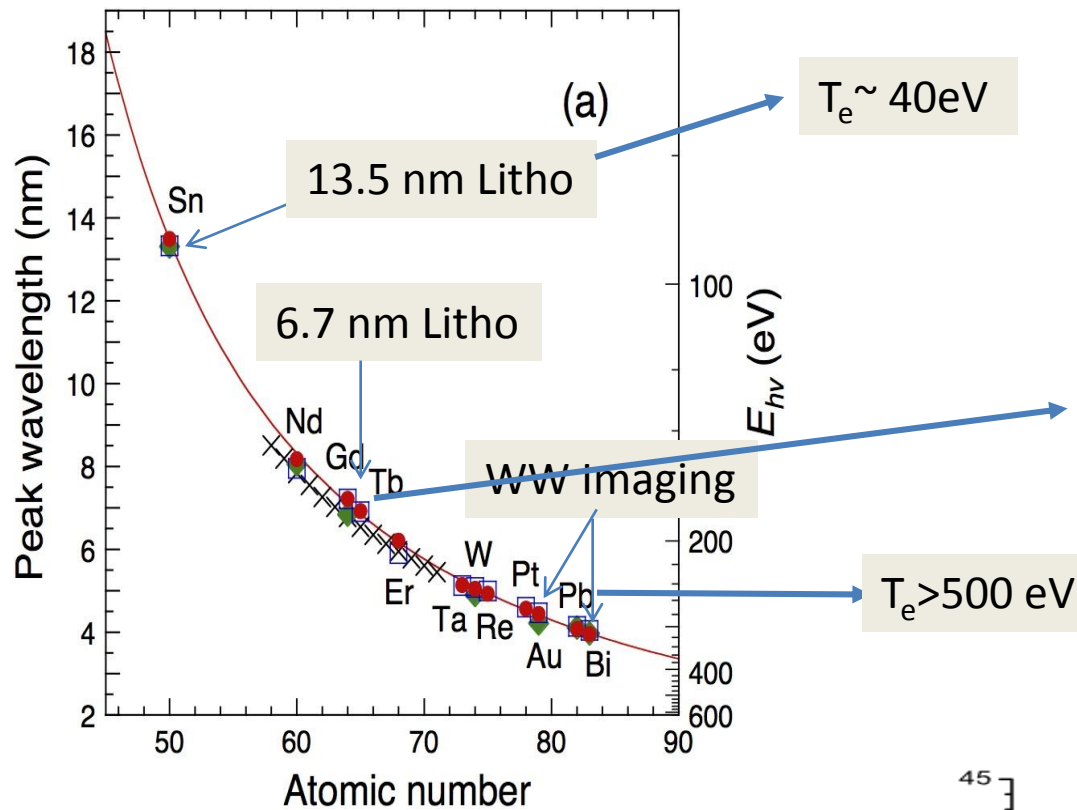
(Mandelbaum et al. Phys. Rev. A35, 5051, 1987)

CI effects (mixing) should diminish as Z increases

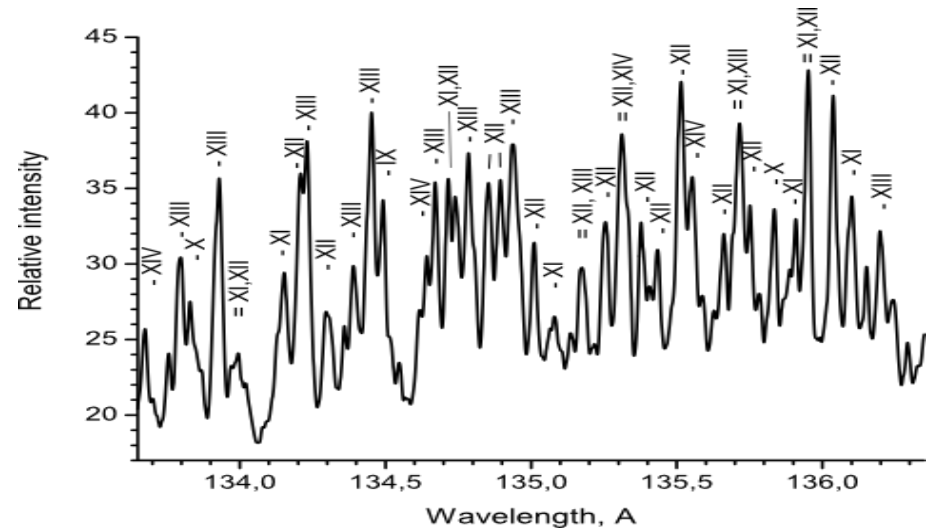
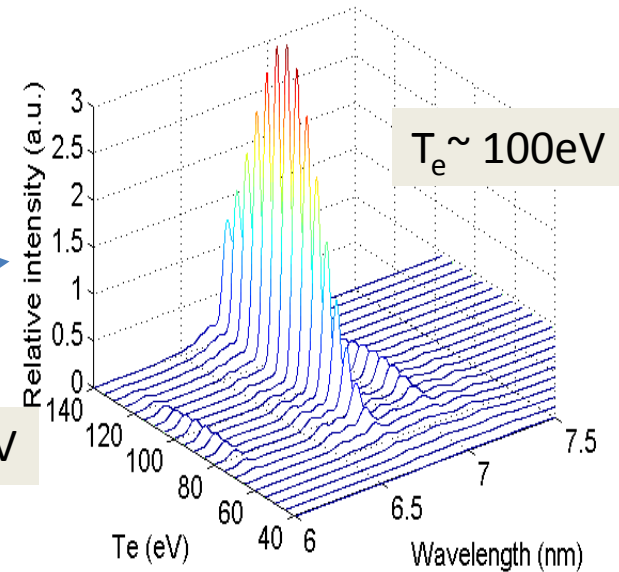


Kilbane and O'Sullivan PRA 82, 062504 (2010)
Kilbane JPB 44 165006 (2011)

Evolution with Z of $\Delta n=0$, $n=4 - n=4$ UTAs

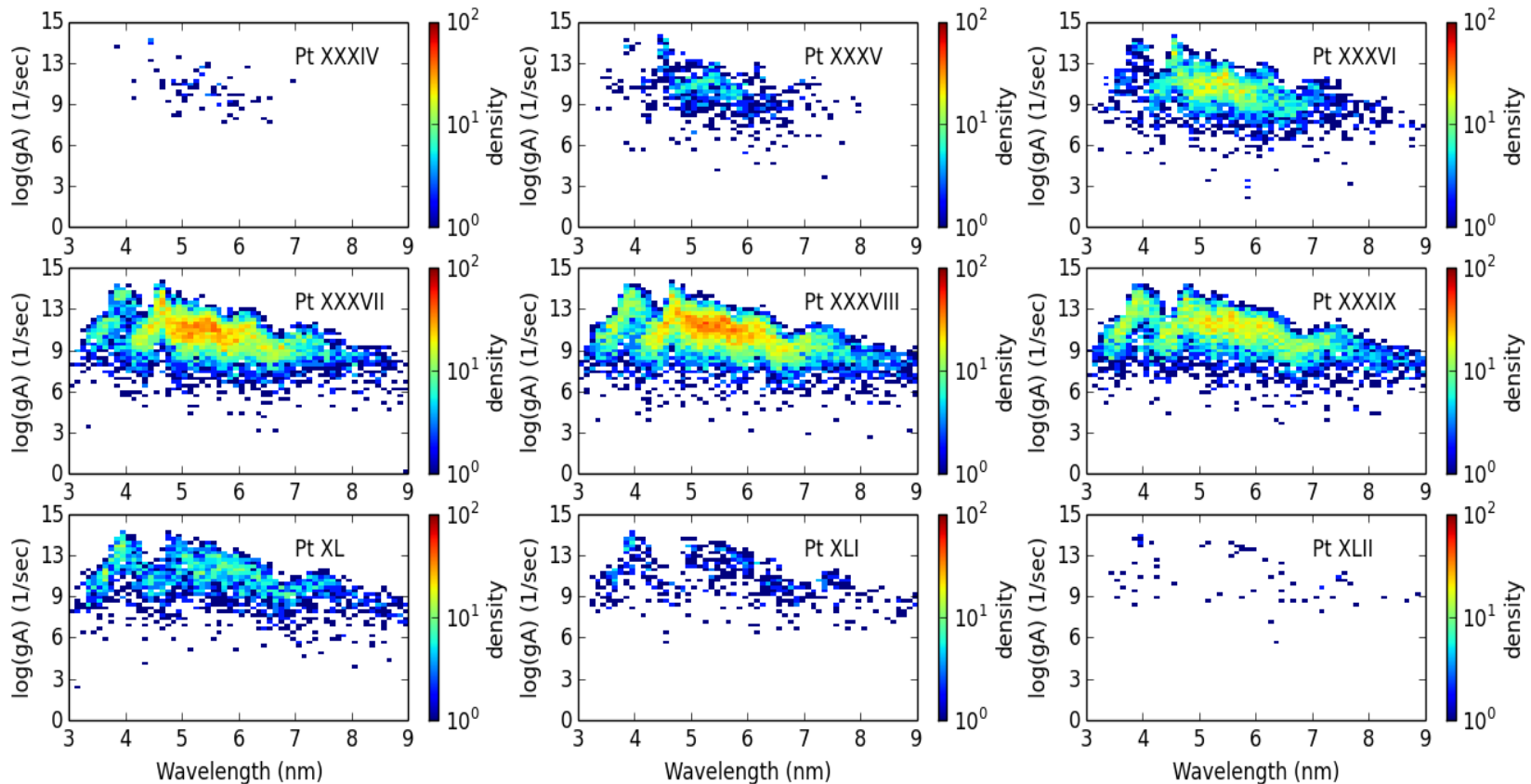


Plasma electron temperature required increases with Z



*Analysis of Sn by Churilov and Ryabtsev
Phys. Scr. 73 614-619, 2006*

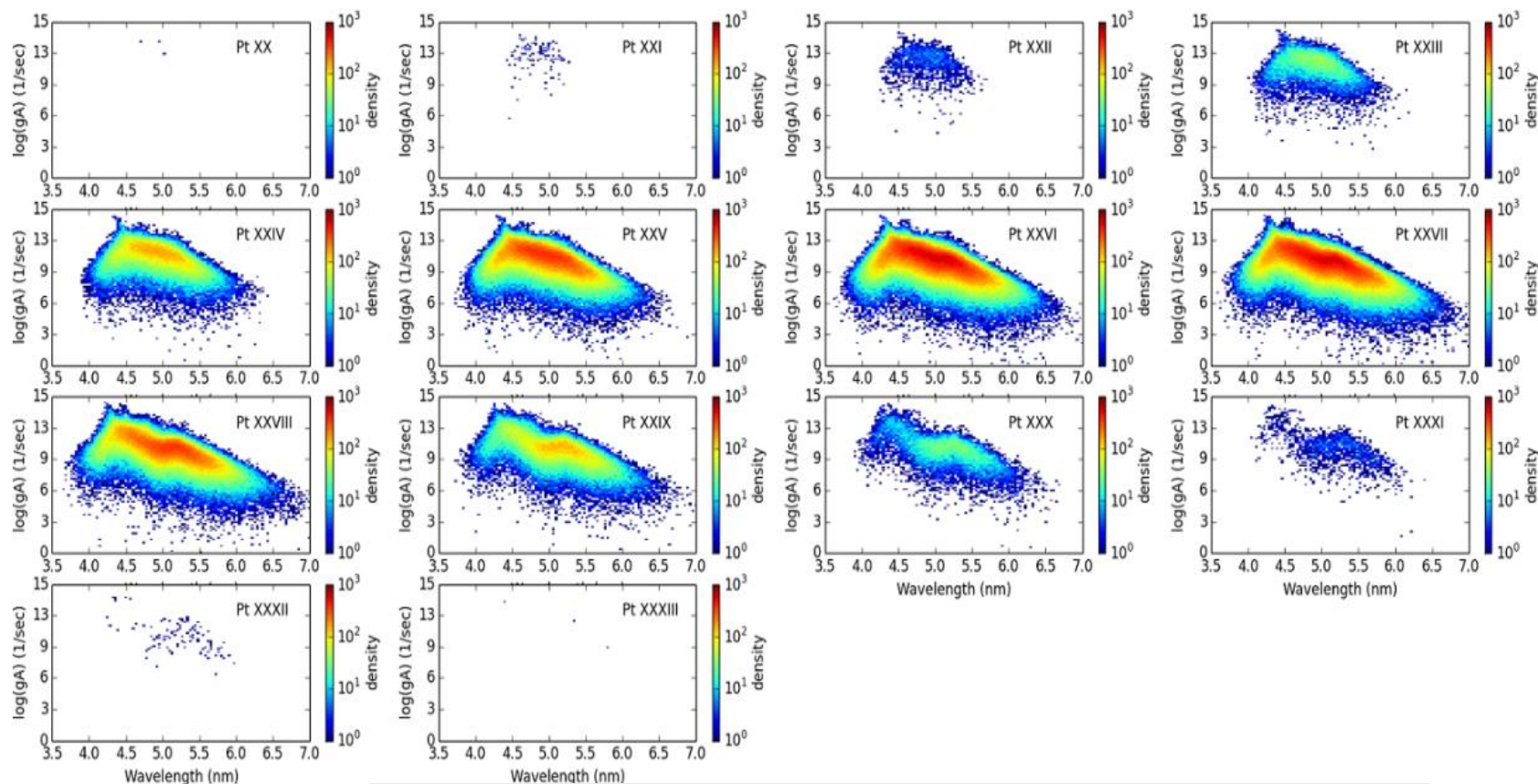
Complexity of $\Delta n=0$ transitions in Pt XXXIV ($4d^9$) to Pt XLII ($4d^1$)



$4p^6 4d^m - 4p^5 4d^{m+1} + 4p^6 4d^{m-1} 4f$ transitions

Additional complexity at higher Z, $4d^{10}4f^N$ - $4d^94f^{N+1}$

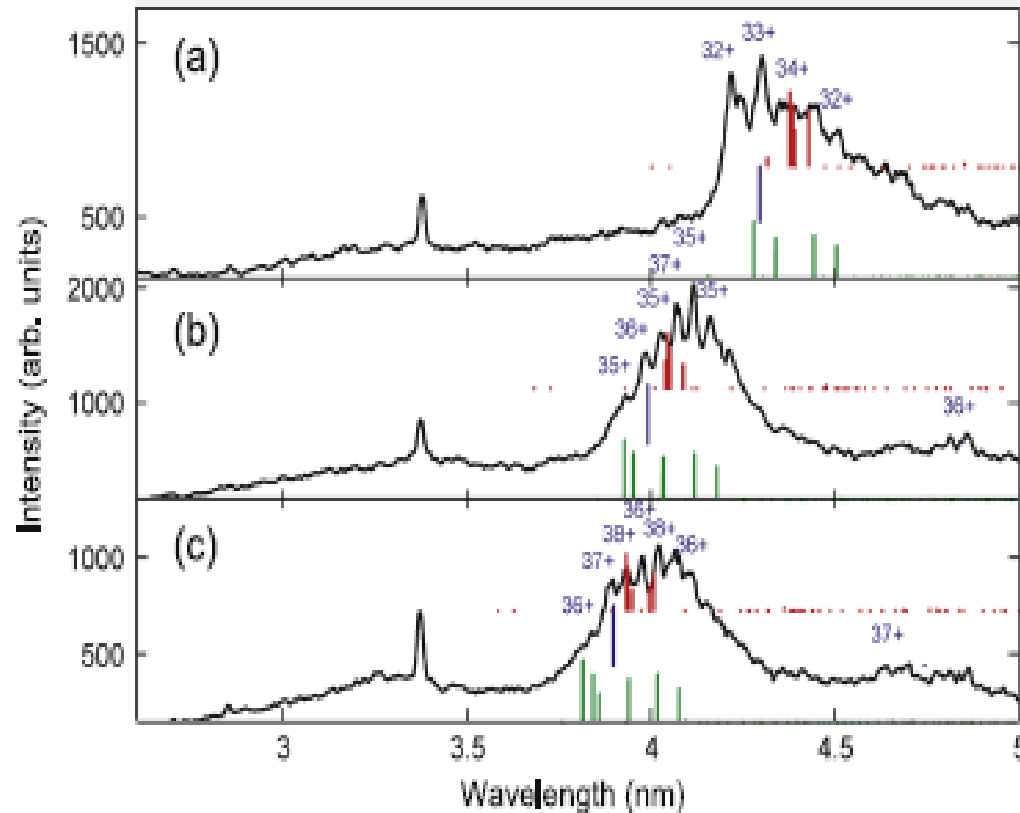
Arrays: Pt XX, (N=13)- Pt XXXIII, (N=0)



The most important transitions occur in Ag-like, Pd-like and Rh-Like Ions with $4d^{10}4f$, $4d^{10}$ and $4d^9$ ground states

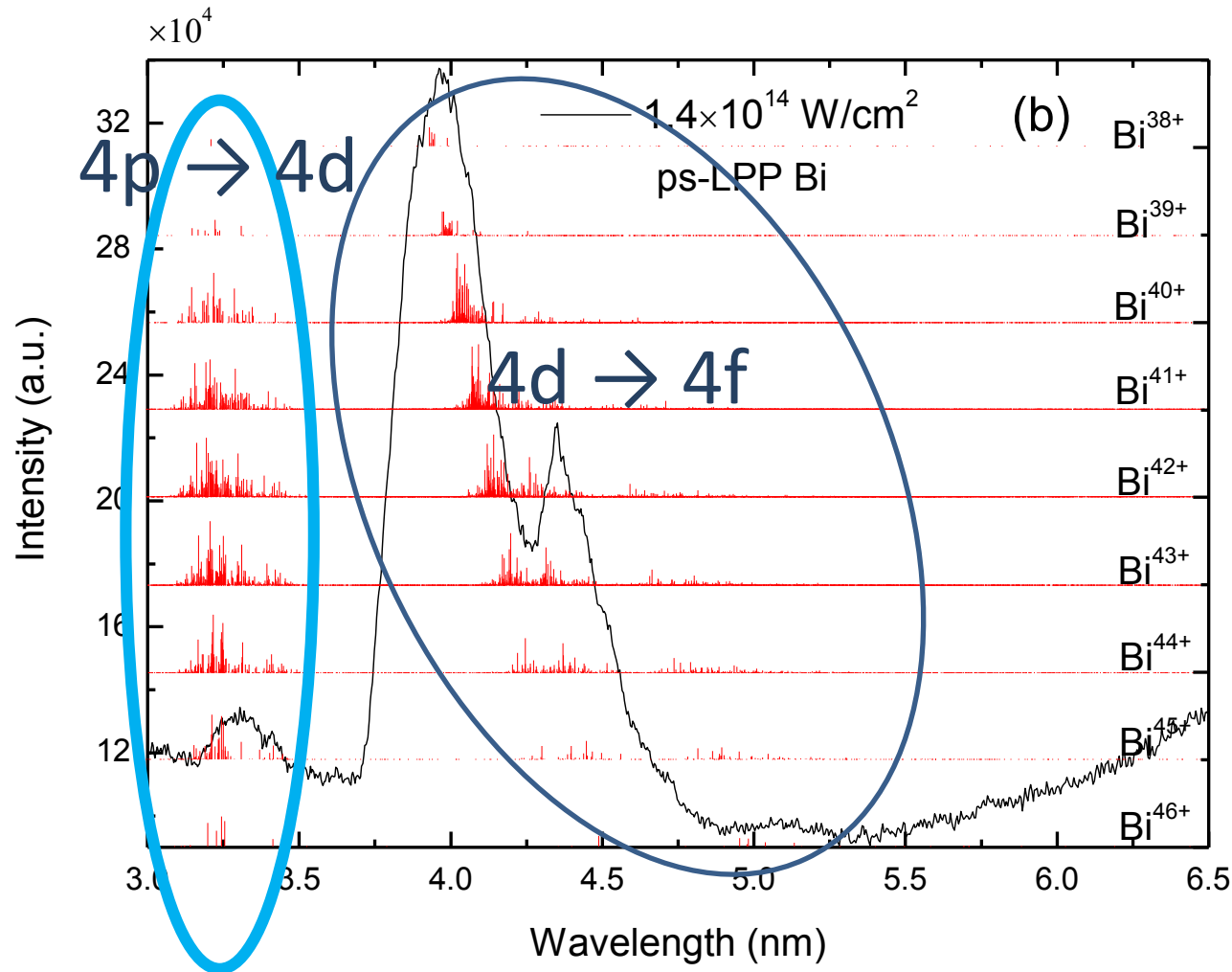
(Sugar and Kaufman Phys. Scr. **24**, 742 (1982) and **26**, 417 (1984))

$\Delta n = 0$ 4-4 UTAs in the water window region, Au, Pb and Bi



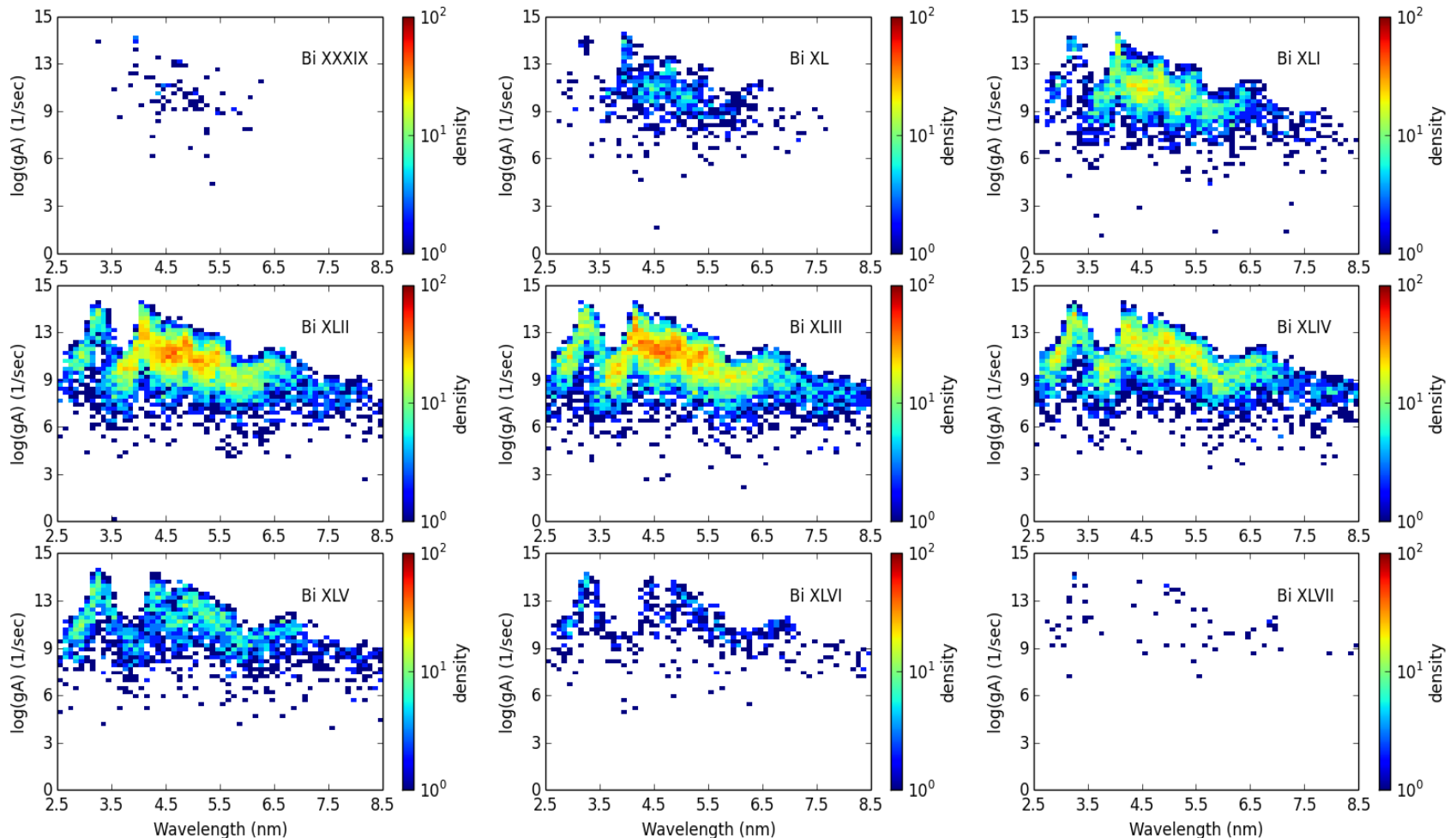
- Au, Pb and Bi injected using LPP or pellet.
- Spectra dominated by resonant line emission to the ground state
- Only Ag-, Pd- and Rh-like ions give line emission
- Absorption free

$\Delta n = 0$ 4-4 UTAs in a Bi LPP; comparison with calculations



4p \rightarrow 4d involve creation of a hole in sub-valence 4p subshell, not strong in emission. More important for absorption. Same probably true for strongest 4d¹⁰4f^N-4d⁹4f^{N+1} lines

Complexity of $\Delta n=0$ transitions in Bi XXXIX ($4d^9$)- Bi XLVII ($4d^1$)



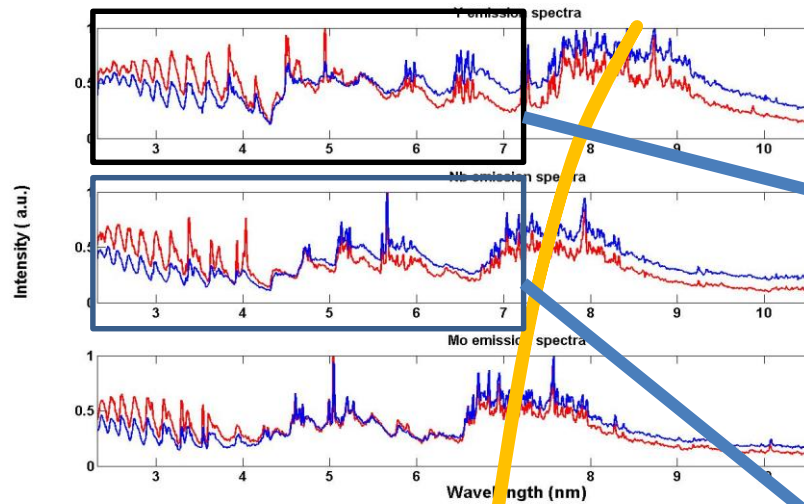
$4p^64d^m-4p^54d^{m+1}+4p^64d^{m-1}4f$ transitions

$\Delta n = 1$ UTAs

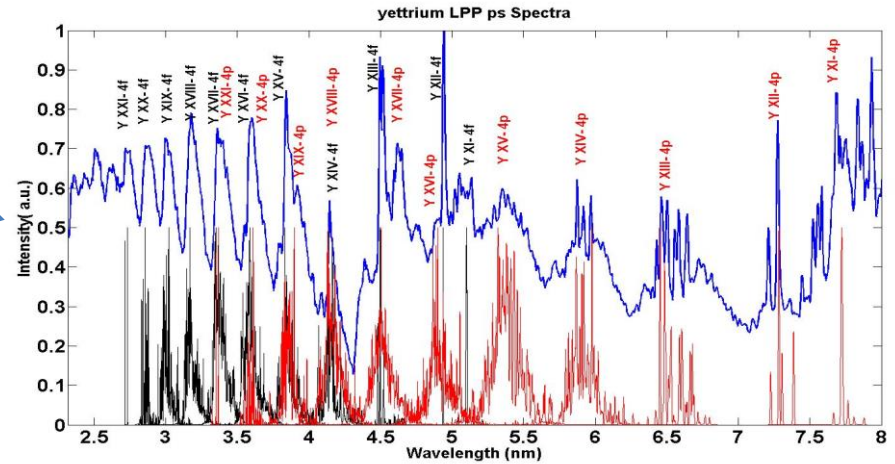
Periodic Table of the Elements

Periodic Table of the Elements																					
<div>Atomic Number</div> <div>Symbol</div> <div>Name</div> <div>Atomic Mass</div>																					
1 1A 1 H Hydrogen 1.008	2 2A 2 He Helium 4.003															5 3A 5 B Boron 10.811	6 4A 6 C Carbon 12.011	7 5A 7 N Nitrogen 14.007	8 6A 8 O Oxygen 15.999	9 7A 9 F Fluorine 18.998	10 8A 10 Ne Neon 20.180
3 Li Lithium 6.941	4 Be Beryllium 9.012															13 Al Aluminum 26.982	14 Si Silicon 28.086	15 P Phosphorus 30.974	16 S Sulfur 32.066	17 Cl Chlorine 35.453	18 Ar Argon 39.948
11 Na Sodium 22.990	12 Mg Magnesium 24.305	3 3B 21 Sc Scandium 44.956	4 4B 22 Ti Titanium 47.88	5 5B 23 V Vanadium 50.942	6 6B 24 Cr Chromium 51.996	7 7B 25 Mn Manganese 54.938	8 VIII 26 Fe Iron 55.933	9 VIII 27 Co Cobalt 58.933	10 VIII 28 Ni Nickel 58.693	11 1B 29 Cu Copper 63.546	12 2B 30 Zn Zinc 65.39	13 Ga Gallium 69.732	14 Ge Germanium 72.61	15 As Arsenic 74.922	16 Se Selenium 78.09	17 Br Bromine 79.904	18 Kr Krypton 84.80				
19 K Potassium 39.098	20 Ca Calcium 40.078	39 Y Yttrium 88.906	40 Zr Zirconium 91.224	41 Nb Niobium 92.906	42 Mo Molybdenum 95.94	43 Tc Technetium 98.907	44 Ru Ruthenium 101.07	45 Rh Rhodium 102.906	46 Pd Palladium 106.42	47 Ag Silver 107.868	48 Cd Cadmium 112.411	49 In Indium 114.818	50 Sn Tin 118.71	51 Sb Antimony 121.760	52 Te Tellurium 127.6	53 I Iodine 126.904	54 Xe Xenon 131.29				
55 Cs Cesium 132.905	56 Ba Barium 137.327	57 La Lanthanum 138.906	58 Ce Cerium 140.115	59 Pr Praseodymium 140.908	60 Nd Neodymium 144.24	61 Pm Promethium 144.913	62 Sm Samarium 150.36	63 Eu Europium 151.966	64 Gd Gadolinium 157.25	65 Tb Terbium 158.925	66 Dy Dysprosium 162.50	67 Ho Holmium 164.930	68 Er Erbium 167.26	69 Tm Thulium 168.934	70 Yb Ytterbium 173.04	71 Lu Lutetium 174.967	86 Rn Radon 222.018				
87 Fr Francium 223.020	88 Ra Radium 226.025	89-103 Actinide Series	104 Rf Rutherfordium [261]	105 Db Dubnium [262]	106 Sg Seaborgium [266]	107 Bh Bohrium [264]	108 Hs Hassium [269]	109 Mt Meitnerium [268]	110 Ds Darmstadtium [269]	111 Rg Roentgenium [272]	112 Cn Copernicium [277]	113 Uut Ununtrium unknown	114 Fl Flerovium [289]	115 Uup Ununpentium unknown	116 Lv Livermorium [298]	117 Uus Ununseptium unknown	118 Uuo Ununoctium unknown				

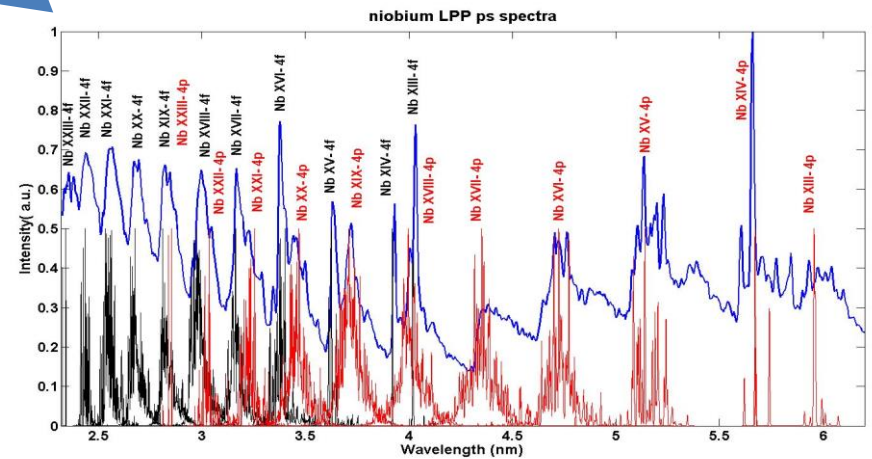
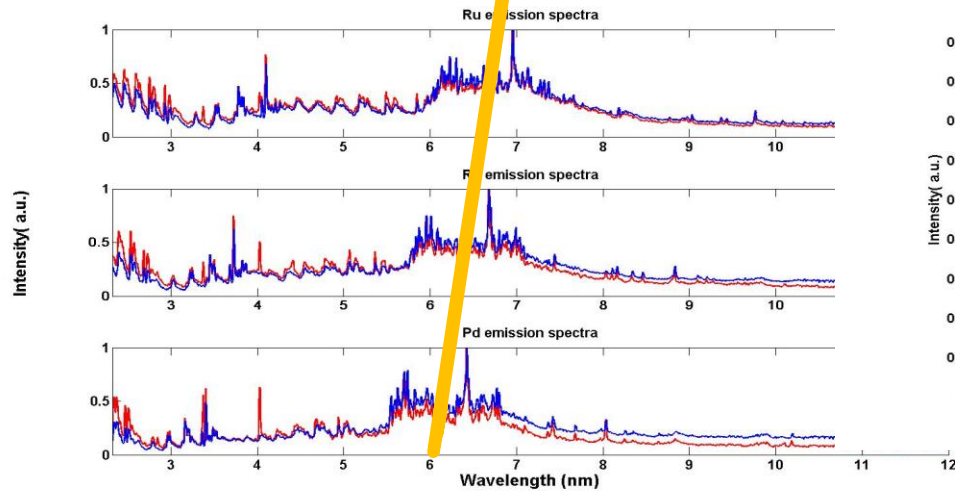
$\Delta n=1$ transitions in 2nd transition row



$\Delta n=0$, 3p-3d



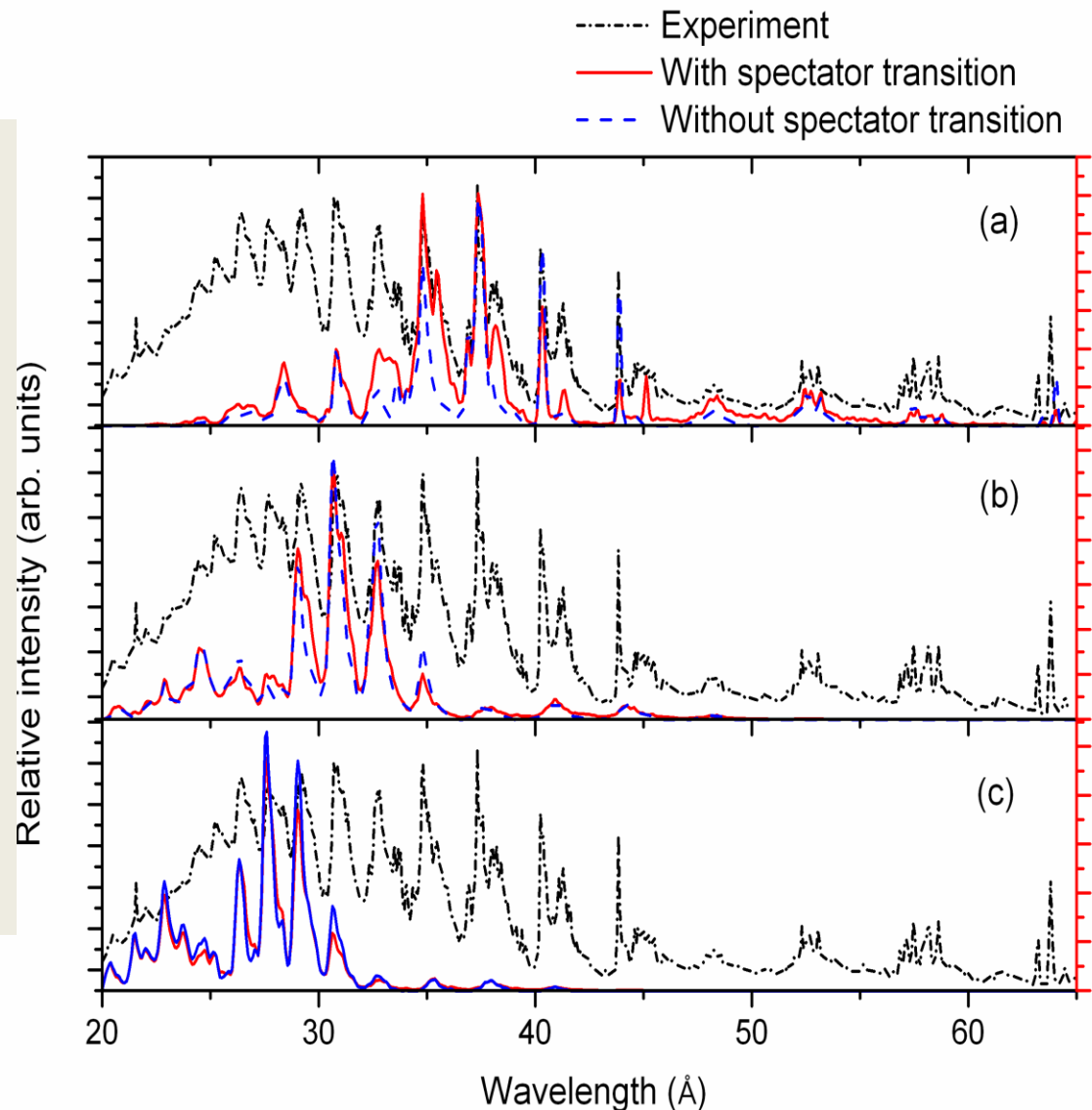
$\Delta n=1$, 3d-4p and 3d-4f



Importance of satellite line emission

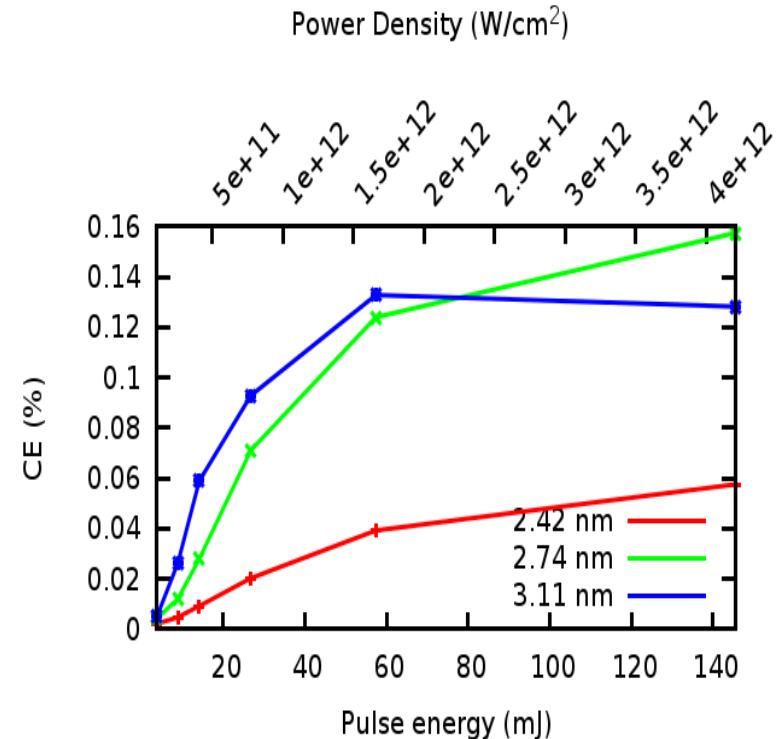
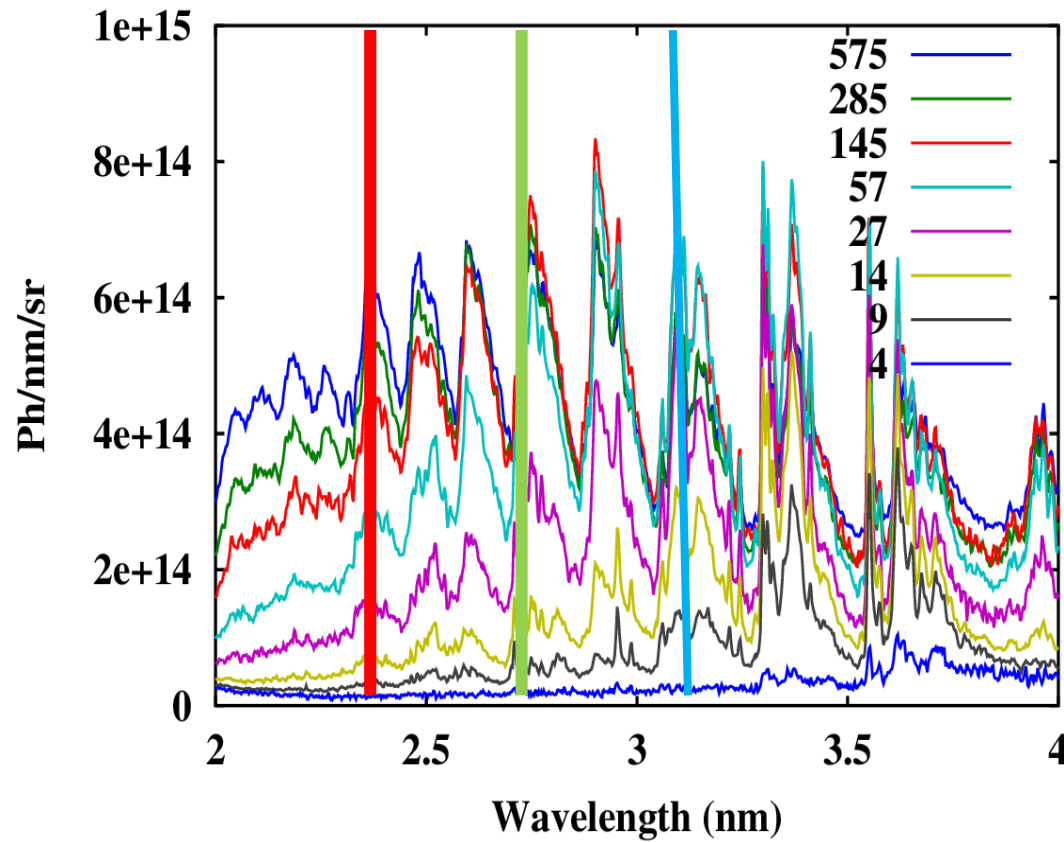
Satellite line emission of type $3d^{n-1}4s-3d^{n-2}4s4f$ very important.

Comparison between the observed spectrum and numerically calculated spectra of Zr with and without satellite lines assuming a steady-state CR model for different plasma temperatures of 100 (a), 150 (b), and 200 eV (c).

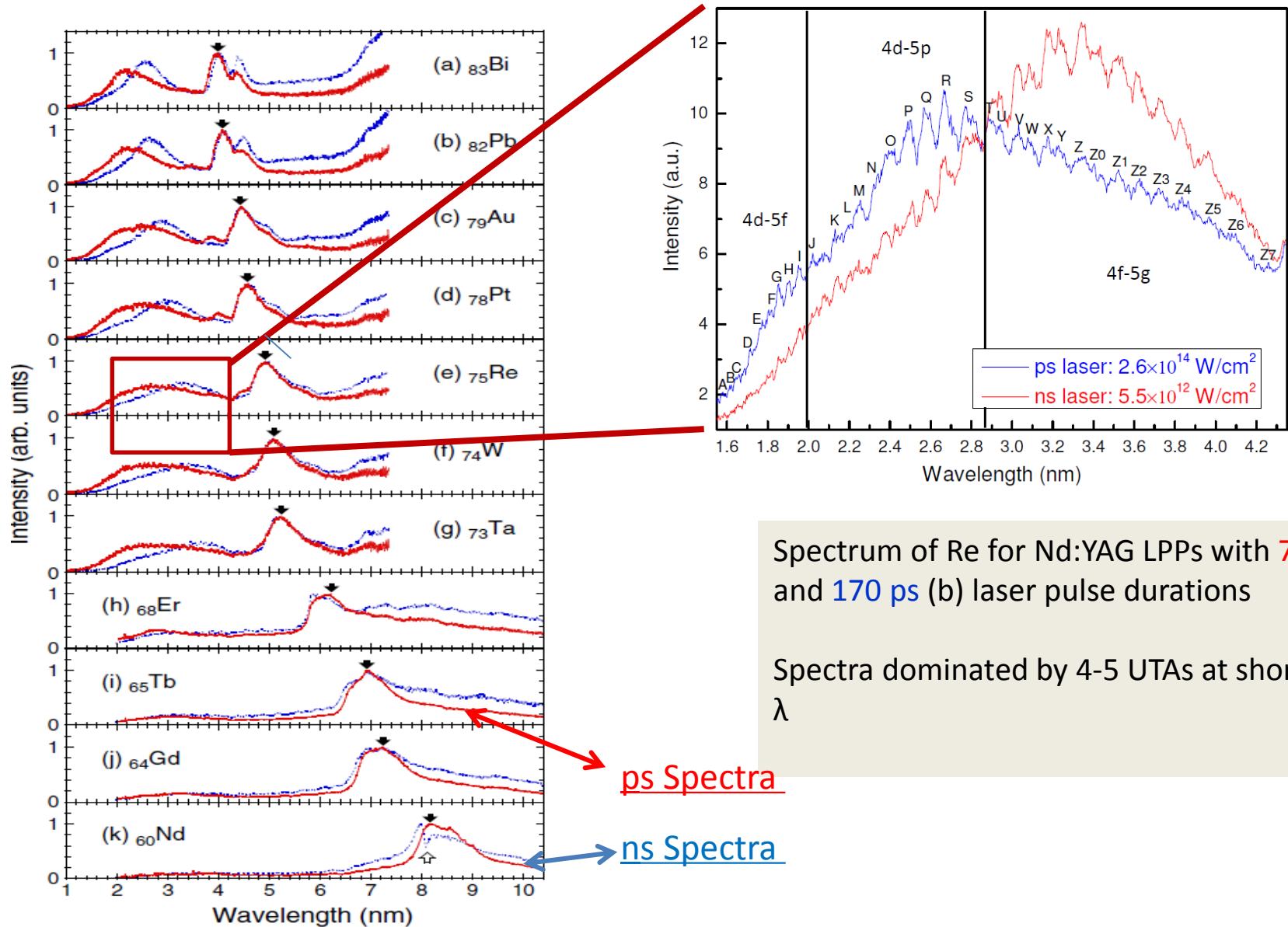


Mo as a WW source: CE vs pulse energy or Φ @ different λ

Energy Normalised Spectra of Mo at Diff Pulse Energies



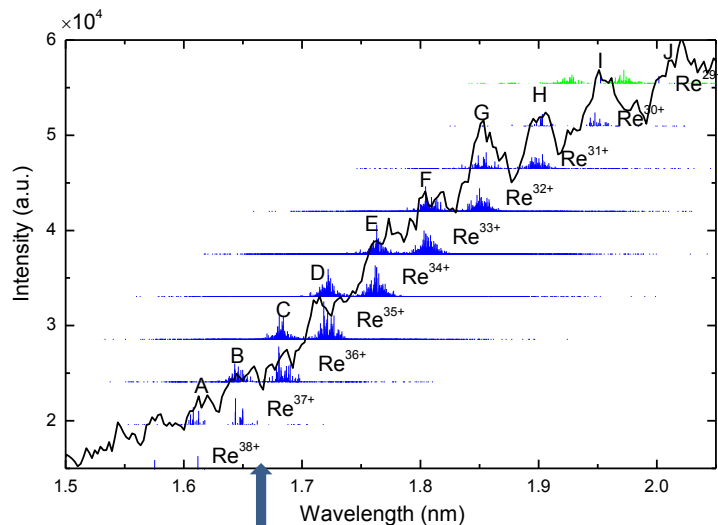
3rd transition row spectra



Spectrum of Re for Nd:YAG LPPs with **7 ns** and **170 ps** (b) laser pulse durations

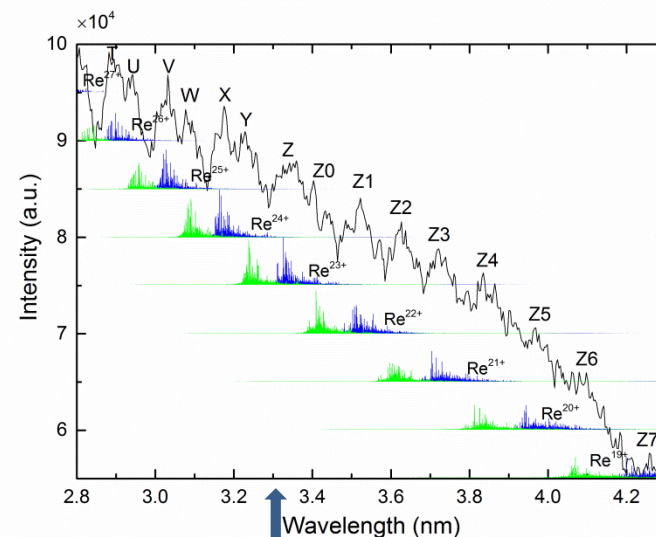
Spectra dominated by 4-5 UTAs at shorter λ

Spectral identification in Re (Z=75)

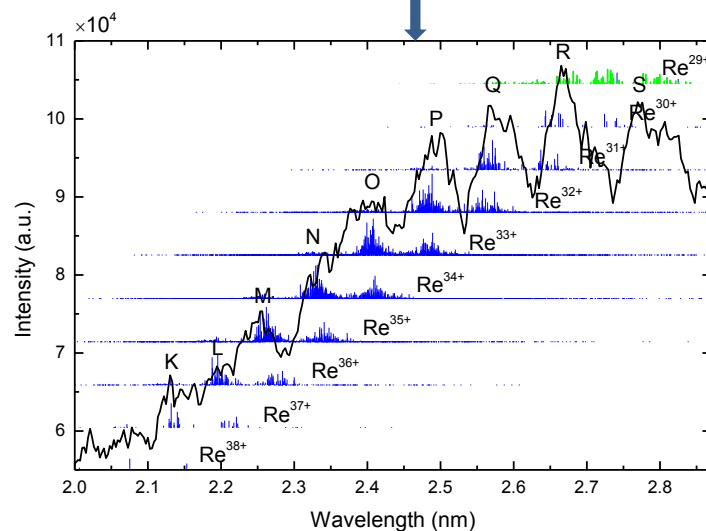


$4d^N-4d^{N-1}5p$ in
 $\text{Re}^{29+}-\text{Re}^{38+}$ and
 $4d^N-15s^1-4d^{N-}$
 $25s^15p$ satellites
 in Re^{29+}

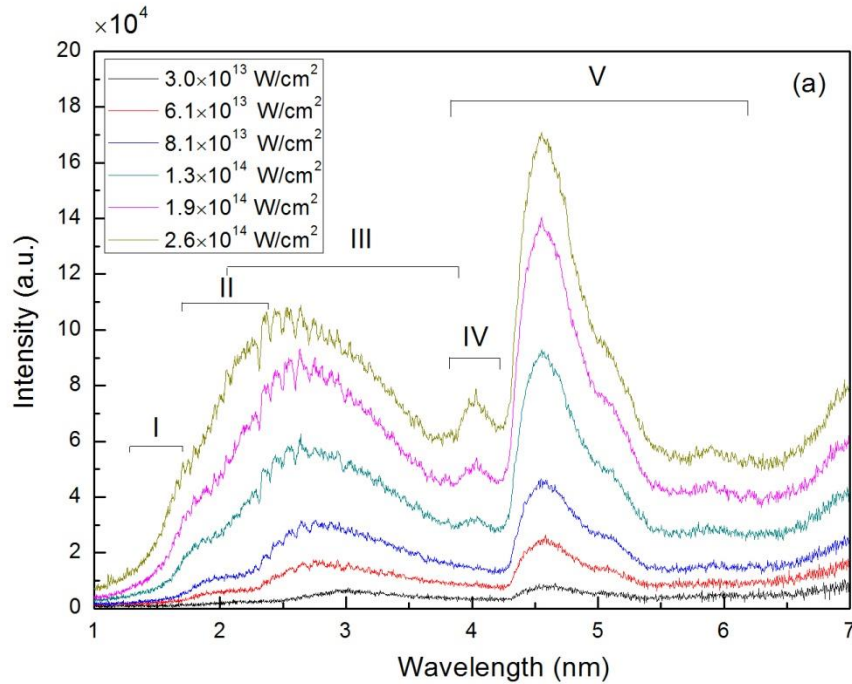
$4d^N-4d^{N-1}5f$ in $\text{Re}^{29+}-$
 Re^{38+} and $4d^N-15s^1-$
 $4d^N-25s^15f$ satellites
 in Re^{29+}



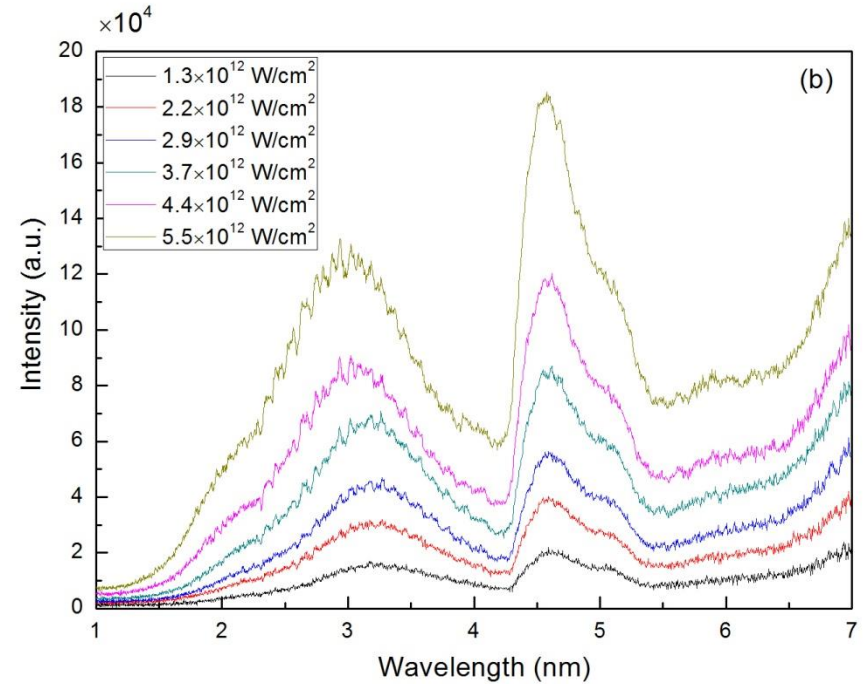
$4f^N-4f^{N-1}5g$ in $\text{Re}^{21+}-\text{Re}^{27+}$
 and $4f^N-15s^1-4f^{N-}25s^15g$
 satellites in $\text{Re}^{19+}-\text{Re}^{27+}$



SXR spectra of Pt (Z=78)



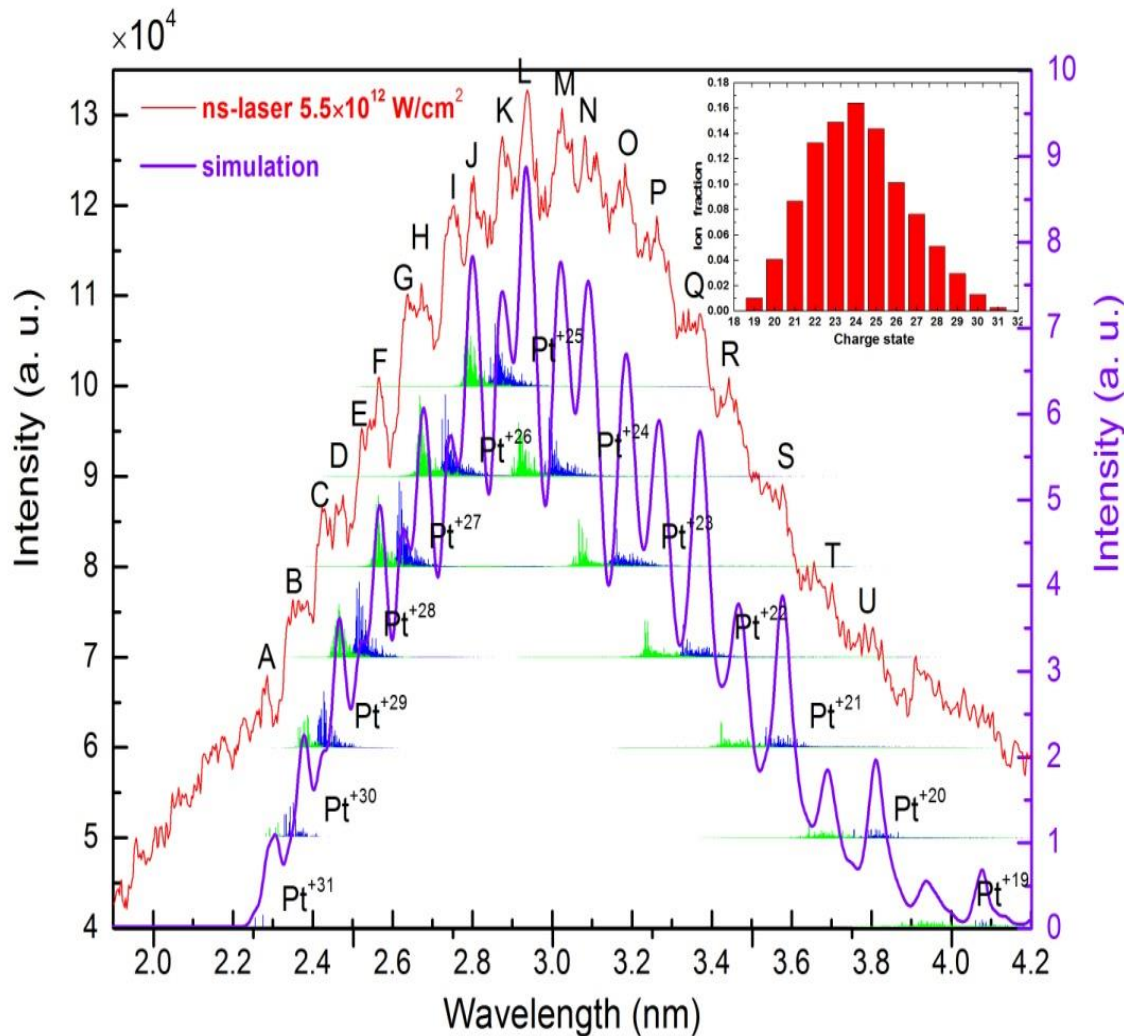
LPP produced by 170 ps Nd:YAG laser



LPP produced by 10 ns Nd:YAG laser

The contributions from different transitions are labelled as follows: (I) $\Delta n=1$, $4d^m-4d^{m-1}5f$; (II) $\Delta n=1$, $4d^m-4d^{m-1}5p$; (III) $\Delta n=1$, $4f^m-4f^{m-1}5g$ and $4f^{m-1}5/-4f^{m-2}5/5g$ ($l=s, p$); (IV) $\Delta n=0$, $4p^64d^m-4p^54d^{m+1}$; (V) $\Delta n=0$, $4p^64d^m-4p^54d^{m+1}+4p^64d^{m-1}4f$ and $4d^{10}4f^m-4d^94f^{m+1}$. (b) Pt spectra from ns-LPP recorded in the wavelength region 1-7 nm at six different power densities.

$\Delta n=1$, 4f-5g Transitions in Pt



Comparison of calculated gA value distributions of $4d^{10}4f^m-4d^{10}4f^{m-1}5g$ and $4d^{10}4f^{m-1}5s-4d^{10}4f^{m-2}5s5g$ transitions in Pt XX-Pt XXXII and a simulated UTA spectrum with a measured Pt spectrum from a Nd:YAG ns-LPP.

The insert shows the ionic populations that fit the spectrum. The theoretical spectra are calculated gA-value distributions.

Plans /Conclusions

- Population information especially from calibrated ps spectra.
- Complete studies of high Z spectra. In particular, 4f-5g transitions in lanthanides and explore 4f,5p and 4f, 5s level crossing effects on the spectral appearance.
- Study emission from short pulse CO₂ plasmas, effects of prepulses.
- $\Delta n=1$ UTA in many elements can be found to match MLM reflectivity peaks. Build up comprehensive overview.
- Implement source in XUV microscope.

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